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Part I

THE ELASTIC PROPERTIES OF MOLLUSCAN SHELLS

1 INTRODUCTION

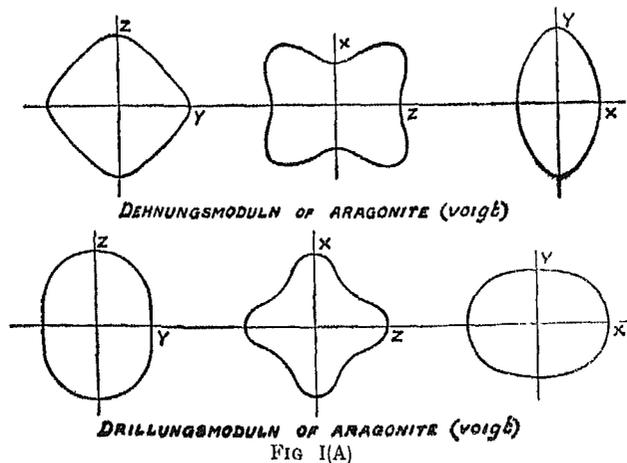
From the theoretical standpoint, measurements of the elastic properties of solids possess the greatest significance when they relate to the material in the form of single crystals, the uniformity of spacing and orientation of the units composing the crystal lattice opens up the possibility of theoretical computations of the elastic properties for comparison with the experimental results. The great majority of the solids used in the arts and industries are, however, not single crystals but aggregates having more or less a complex structure. The relation between the elastic properties of these aggregates and those of the crystalline units of which they are made up is obviously a subject of considerable importance. It may naturally be assumed that the properties of polycrystalline aggregates will depend to a considerable extent on those of the single crystals which form them. The order and arrangement of the crystallites are obviously of importance in this connection. Thus a random orientation of intrinsically anisotropic crystals may be expected to result in an elastically isotropic body while an orientation about a linear axis will result in a fibre structure and consequent elastic anisotropy. In many substances, these considerations are further complicated by the presence of a substance usually of the nature of a cementing material which helps to bind the various crystallites together. The nature, quantity and distribution of this cementing material will influence in a marked manner, the elastic properties of the main substance. This cementing material may, in some cases, be an amorphous phase of the same chemical nature as the crystallites or it may be an entirely foreign substance.

The latter type of cementing material occurs in the molluscan shells which form the subject of the present investigation. There are some varieties of molluscan shells in which the former kind of cementing material occurs. In choosing the materials for this investigation, two considerations have been mainly taken into account. First only such shells have been chosen which are available in large

well-formed shapes so that elastic measurements can be made with accuracy and which form representative types of the different families of molluscan shells. Secondly, the choice has been with a view to include only such shells as are commonly employed for decorative and artistic workmanship. From the point of view of the external appearance, the shells that have been chosen for study can be classified into two kinds, the iridescent and the non-iridescent shells. As is evident from the description, the iridescent shells, on being polished, present a very beautiful array of surface colours which change with changing angles of observation while the non-iridescent shells, on polishing, present only a bright white surface, more or less of the nature of porcelain. The crystalline part of these shells consists of crystals of calcium carbonate and amounts to as much as 85 to 95% (per cent) of the weight of the shells. These crystals of calcium carbonate are held together in the frame-work of the shells by an organic protein matter called conchyolin. Chalk, as is well-known, occurs in nature in two distinctly crystalline varieties known as aragonite and calcite. It is the former variety of it which occurs in the iridescent shells commonly called mother-of-pearl while the latter variety occurs in the non-iridescent shells.

A detailed experimental study of the elastic properties of calcite and aragonite has been made by Voigt (1890 & 1907) Fig I(A)

ELASTIC PROPERTIES



gives the principal sections, according to Voigt, of the elastic surfaces of aragonite and Fig I(B) those of calcite. The striking

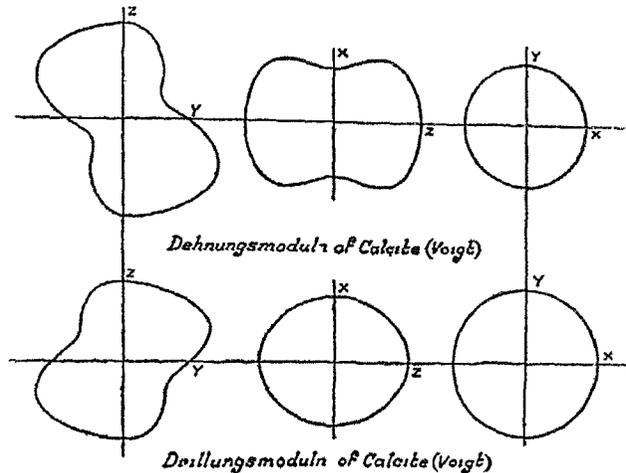


FIG I(B)

difference in elastic properties between the two forms is obvious from the figures. Oke (1936) has made a theoretical computation of the elastic constants of aragonite and calcite based on Born's Lattice Theory of Crystal structure and finds a satisfactory agreement between his calculations and the experimental values of Voigt. According to Oke, the cause of the difference in the elastic properties between calcite and aragonite is to be traced to the difference in the relative positions of the Ca and CO₃ ions in the two crystals. The elastic anisotropy is more marked in the case of aragonite than in calcite. Indeed, in the X-Y-plane while calcite is isotropic, the Young's modulus of aragonite along the X-axis is quite double that along the Y-axis. As we shall see later, this feature makes itself felt in the elastic properties of mother-of-pearl.

STRUCTURE AND CONSTITUTION OF THE SHELLS

2 STRUCTURE OF MOTHER-OF-PEARL

Dating from the time of Brewster (1853), the structure of mother-of-pearl has formed the subject of numerous investigations, amongst which those of Schmidt (1924) should be specially mentioned. Boggild (1930) has made a detailed study of the architecture of

molluscan shells by means of the polarising microscope succeeded in classifying them into definite groups like homogeneous structure, the laminated or the nacreous stratified structure, the twinned laminated structure, the fibrous structure and so on. As a result of these studies, it is found that mother-of-pearl has got a laminated structure, consisting of alternate layers of aragonite and conchyolin. Each aragonite layer is made up of a number of platelets of aragonite, the platelets being bound to one another by intervening protein layers. According to Schmidt and as confirmed by Boggild, the platelets forming the platelets in all the iridescent shells have their C-axes normal to the plane of the platelets, which is parallel to the surface of the shells. Schmidt had also determined the thickness of the protein layers intervening between the aragonite layers, which was small in comparison with the thickness of the latter, a result also arrived at by Raman (1935) by a study of lamellar interference light by the surface of these shells. By a study of the interference and diffusion haloes exhibited by these shells, Raman established that the platelets of aragonite forming the shell are sensibly of uniform dimensions and spaced in a regular manner. The three Great groups of molluscs, Bivalves, the Gastropods and the Cephalopods are characterized by striking differences in their diffraction haloes which indicate that the arrangements of the crystalline particles are very different in the groups. He thus finds that in the bivalves there is a more or less regular orientation of the aragonite crystals with respect to the lines of growth in the plane of the shell, a regular orientation in the Gastropods and an intermediate orientation in the Cephalopods. From the angular size and general character of the diffraction haloes Raman has been able to estimate the size and disposition of the crystalline particles. An X-ray investigation of the nacreous shells has enabled Ramaswami (1935) to confirm the conclusions drawn from these optical observations and to establish the orientation of the crystals in the different shells. He finds that in the shells examined the c-axis is normal to the shell-s

the a and b-axes lying in the plane of the shell have got a regular orientation (with only an error of 5°) with respect to the lines of growth in the Lamellibranchs and a quite random orientation in the Gastropods. In 'Nautilus Pompilius' belonging to the Cephalopod group, a preferred orientation is complicated with twinning and large error of orientation of about 15° . Ramaswami's observations on Nautilus are of special interest from the viewpoint of the present investigation and are referred to in detail later on. Differences between the diffraction haloes of shells within the same group observed by Raman are correlated with variations in the size of the platelets of aragonite and a varying amount of error in their orientation. In observations with parallel and convergent polarised light under the petrographic microscope, Rajagopalan (1936) has made measurements of the size and arrangement of the crystal particles in the various shells and finds confirmation of the previous observations. In the present investigation, the elastic properties of nacre of different shells have been studied and it is shown that the relation between the elastic properties of aragonite and the shells is intelligible in view of the structural details established by previous investigators. As shown later, it has also been possible to obtain a quantitative estimate of the protein distribution in nacre.

B STRUCTURE OF NON-IRIDESCENT SHELLS

Of the two calcareous shells studied in this investigation, the 'chank' belongs to the Gastropod group while the 'Placuna Placenta' commonly known as the window-pane oyster belongs to the Bivalve group. The shell of the chank possesses what Boggild calls the homogeneous structure, appearing like a piece of porcelain without any details of structure when examined under ordinary light. When examined between crossed nicols, however, definite extinction is observed in specific directions, which in this shell, exhibits a certain amount of error of orientation. The structure of the window-pane oyster shell is what is called the foliated structure, the shell being made up of a bundle of almost parallel leaves. The shell architecture is distinctly non-homogeneous easily flaking off in one direction and tearing

pieces in another. Under the polarising microscope, the optic axis definitely shows an inclination to the shell-surface deviating appreciably from 90° but possessing less error of orientation than the chank. This kind of foliated structure is the calcitic analogue of the nacreous structure of aragonite as it occurs in mother-of-pearl. By studying the magnetic anisotropy of these shells, Nilakantan (1937), has established the angle of orientation of the optic axis to the shell-surface. The crystallites of calcite form a brick-work like structure, the bricks being rather disproportionately elongated and cemented together by a layer of conchyolin all round. There is a rather liberal use of this cementing material in this shell and as in the case of 'M Margaritifera,' it has been possible to obtain a quantitative estimate of the distribution of the cementing material from considerations of the observed elastic modulus of the shell.

C. THE CHEMICAL COMPOSITION OF THE SHELLS

The relative abundance of crystalline and cementing matter in the architecture of the shell will exercise a large control on the ultimate strength of the shell and its elastic behaviour and so it was considered necessary to make a chemical analysis of the shells in order to determine the percentage of the constituents. The method of analysis is particularly simple since it is known that only two substances viz, calcium carbonate and conchyolin make up the shell. The analysis was done by two independent methods, in one of which the calcium was estimated as oxide and in the other as carbonate. Having determined the amount of calcium carbonate, the remainder was taken as conchyolin. The specimens that had been employed in the determination of the elastic moduli were first dried for about an hour in an air oven maintained at 102° and then reduced to a fine powder in an agate mortar. This powder, which was kept inside a desiccator till actually required for the analysis, served as the starting material for the methods of analysis. In the first method a known amount of the powder was calcined to constant weight at bright red heat in a crucible and the mass of the oxide thus obtained was determined. The equivalent amount of carbonate was calculated

and the balance was taken as conchyolin. In the second method, a known amount of shell powder was dissolved in excess of dilute hydrochloric acid and the calcium was precipitated as oxalate in an ammoniacal medium. The precipitate, carefully washed and collected in a crucible was converted into the carbonate by heating to a dull-red heat with the usual precautions and the carbonate obtained from the known amount of shell was determined, the balance being taken as conchyolin.

The results of the chemical analysis are given in the following table.

TABLE I
Chemical Composition of the Shells

Name of Shell	Percentage of calcium carbonate		Percentage of conchyolin	
	I method	II method	I method	II method
1 M Margaritifera .	95.8	95.0	4.2	5.0
2 M Vulgatus .	90.0	89.0	10.0	11.0
3 Trochus	97.5	97.0	2.5	3.0
4 Turbo ...	93.5	92.5	6.5	7.5
5 Helotis .	92.0	91.0	8.0	9.0
6 Nautilus Pompilius .	86.0	85.0	14.0	15.0
7 Window-pane oyster .	90.2	89.7	9.8	10.3
8 Chank ..	92.8	92.4	7.2	7.6

3 PREPARATION OF THE SPECIMENS

In Sir C V Raman's private collection of shells, large and well-formed shells were available belonging to the various families and the following table gives details of the shells that were employed in the present investigation

TABLE II
Description of the Shells studied

No	Family	Name of Shell	Crystalline Component	Source	Approximate Dimensions
1	Lamellibranchs	M Margaritifera	Aragonite	Bombay market	8" x 6"
2	"	M Vulgaris	"	Rameswaram (Madras)	2 5" x 1 8"
3	"	Placuna Placenta	Calcite	Bombay market	6" x 5"
4	Gastropods	Turbo	Aragonite	"	6" diameter 5" Height
5	"	Trochus	"	Andamans	5" diameter 4" Height
6	"	Helix (Abalone C)	"	California	8" x 5"
7	"	Turbinella Pium (Indian chank)	Calcite	Rameswaram (Madras)	4" diameter 6" Height
8	Cephalopods	Nautilus Pompilus	Aragonite	Ennur (Madras)	6" diameter 3" Height

As will be seen from the above table except 'M Vulgaris' of which a specimen larger than about 2" was not available, all the other shells were quite large specimens and it was not a difficult matter to prepare a few good test pieces for measuring the elastic constants. Fig 1 represents the general appearance of a specimen of 'M. Margaritifera' after the outer prismatic layer has been cleaned up by a rapid application of dilute hydrochloric acid. The lines of growth are clearly discernible as large curved lines running nearly parallel to

one another and it is easy to choose a place where the lines are sensibly straight over distances of 3 or 4 cm and in such places a

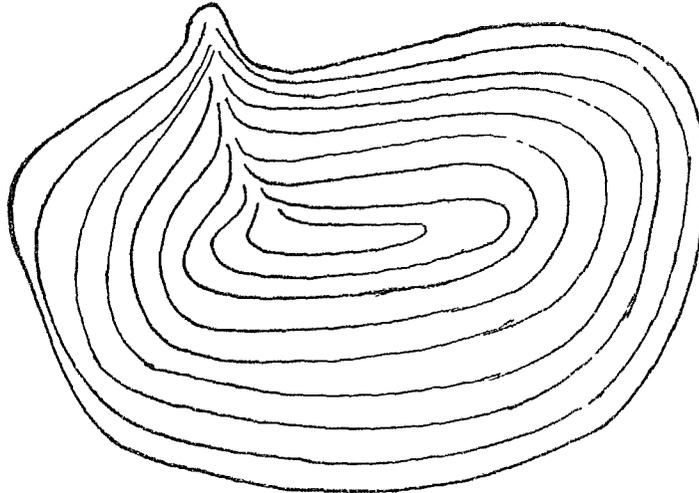


FIG II

strip of about 5 mm width and as great a length as possible is marked out with its length inclined at a known definite angle to the lines of growth. The strip is afterwards carefully cut out with a jeweller's saw and ground on a thick glass sheet with emery powder of increasing fineness so as to remove the outer prismatic layer completely and polish the nacreous layer sufficiently smooth. The characteristic iridescent reflection helps to ensure during the grinding that the surface of the specimen is parallel to the lamination planes. Thus out of the strip cut out, a specimen of about 3 cm length, 5 mm width and 1 mm thickness is carefully prepared. The final grinding is done with the finest grade emery powder and the dimensions are checked at different places with a screw gauge or vernier calipers to within half a per cent deviation from the mean.

For getting transverse sections a specially thick large shell of '*M. Margaritifera*' whose mean thickness was about 5 mm was chosen and at its thickest portion, a strip normal to the shell surface was cut off in a direction parallel to the lines of growth. The laminations are clearly discernible as lines running parallel to the surface of the shell. This strip is first ground so as to have a thick-

ness of about 2 mm i.e., in the direction perpendicular to growth in the plane of the shell. In favourable positions thickness of the shell was as much as 7 or 8 mm. A specimen with length along 01 at a definite known inclination to the C-axis is carefully polished. The width of the specimen was perpendicular to the direction of the lines of growth and had a magnitude of about 4 mm. The thickness in the plane of the shell at right angles to the lines of growth was of the order of a mm or less in most specimens.

In the case of 'Nautilus', though large shells were used in the experiment, on account of the very large curvature and thickness of the shell, specimens of only about 15–20 mm length could be prepared. Further, since it was found that the modulus showed sudden changes from direction to direction, considerable worthwhile checking up the values obtained with specimens obtained from a curved shell and it will be seen from tables where the results for both the shells are given that within the limits of experiment the results are comparable. In 'Trochus' a difficulty was experienced in getting the final surface of the test-piece parallel to the lamination planes. This shell has got a steep spiral structure, there is a large skew angle between the surface of the shell and the lamination planes. Further, the iridescence of this shell is not quite uniform which adds to the difficulty of getting a properly ground surface. Though great care was of course taken in the preparation of specimens, it is doubtful in the case of 'Trochus' whether the desired success was achieved in making the surface parallel to the laminae.

4 EXPERIMENTAL ARRANGEMENT

(1) *Young's Modulus*—(a) For determining the modulus of specimens of at least 2 cm length, Koenig's method of bending was adopted. Two adjustable robust knife-edges were bolted on to a heavy lathe-bed and the specimen was supported between the two knife-edges. Two small plane mirrors were fixed to the upper surface of the specimen with their planes as nearly

dicular to the specimen surface as possible and their reflecting surfaces facing each other. At a distance of about a metre from one of the mirrors was supported a vertical mm scale which was strongly illuminated with an electric lamp close at hand. On the side of the apparatus remote from the scale was mounted a telescope for viewing the image of the scale as reflected by the two mirrors. A stirrup carried on a third light and small knife-edge, resting across the specimen exactly midway between the two supporting knife-edges carried a light scalepan. Care was taken to see that two mirrors were arranged symmetrically with respect to the two knife-edges and the distance apart between them was equal to or slightly greater than that between the two knife-edges. For the same shell the experiment was repeated with different distances between the knife-edges and with samples of different thicknesses. The results justified the anticipation that within limits, the elastic modulus would be independent of the dimensions of the specimen. In calculating the value of the modulus from the observed displacement in the reading of the telescope, the following formula was employed

$$q = \frac{3wl^2(2D+r)}{2bd^3r},$$

where q is the Young's modulus of the material, w is the weight applied at the midpoint, D is the distance between the scale and the mirror facing it, r is the distance between the two mirrors, l is the distance between the two knife-edges, b is the width of the specimen, d is the thickness of the specimen, and r is the observed displacement of the scale reading.

(b) For determining the Young's modulus of specimens of 1 cm length or thereabouts, a single cantilever method of bending was employed. The specimen was firmly clamped horizontally at one end to a stiff short vertical pillar bolted on to the lathe-bed and was loaded by means of a scalepan suspended from a stirrup resting on the specimen near its free end. A thin strip of plane mirror attached vertically to the free end of the specimen and a telescope and scale mounted at about a metre in front were employed to observe the

deflection of the specimen. The modulus was calculated by the formula,

$$g = \frac{12 w l^2 D}{b d^3 r}$$

where l represents the distance on the specimen between the clamped end and the stirrup carrying the scale-pan and the other symbols signify as in the previous formula.

A test experiment conducted on the same specimen by methods (a) and (b) showed that the results obtained were comparable.

(2) *Rigidity Modulus* — For determining the rigidity modulus the following arrangement was found convenient. A stout vertical metallic pillar about $20 \times 2 \times 2$ cm was bolted rigidly to the lathe-bed and carried two cross-arms. The lower cross-arm was a fixed one while the upper one was capable of being clamped anywhere along the pillar. The specimen was, at its upper extremity, firmly clamped to the upper arm and at its lower extremity was clamped axially to a disc. A stout, short, smooth pin attached axially to the lower face of the disc was passed just freely through a hole in the lower arm vertically below the upper clamp and served to prevent lateral oscillations of the specimen while, however, allowing free rotation. A piece of fine silk thread was doubled round a pin fixed on the rim of the disc and its two extremities left the disc tangentially at opposite ends of a diameter and passing over two smooth ball-bearing pulleys, carried light scalepans at the ends. Two small plane mirrors carried on very narrow metallic strips were fixed at a suitable distance apart on the specimen and a telescope and a scale were arranged at a distance of about a metre in front of the mirrors. Since the vertical distance between the two mirrors was of the order of 2 cm or less, it was possible to get the images of the scale as reflected by the two mirrors simultaneously in focus in the field of view of the telescope. When equal weights were added to the two scalepans, the lower end of the specimen was rotated relative to the upper end and the twist varying as the distance from the clamped end produced different angles of rotation of the two mirrors and

corresponding different changes in the telescope reading. The readings of the telescope thus helped to get the relative angle of twist between the two sections and the rigidity modulus, n , was calculated by means of the formula,

$$n = \frac{32 Mgdli}{ab^3 \left[\frac{16}{3} - 3 \frac{b}{a} \right] (r_1 - r_2)},$$

where M represents the mass at each end of the string, g is the acceleration due to gravity, d is the diameter of the disc, l is the distance between the two mirrors, i is the distance between the scale and the mirrors, a is the width of the specimen, b is the thickness of the specimen, and r_1 and r_2 are the observed displacements of the readings from the two mirrors.

This formula is applicable only to specimens whose width is at least six times its thickness and care was taken to see that all the specimens employed satisfied this condition. Further since specimens less than 2 cm in length could not be satisfactorily used in the apparatus, the rigidity modulus in such cases has not been determined.

RESULTS

The following tables give the results of the experiments on the various shells. The results are also represented graphically on polar coordinates so that the radius vector in any direction gives the modulus along that direction, the x-axis being chosen to represent the direction of the lines of growth. For purposes of comparison with Voigt's curves, in the case of some specimens, the values of the extensibility and deformability are plotted instead of Young's modulus and rigidity modulus. The reciprocal of the Young's modulus expressed in grams weight per square millimeter is termed the extensibility and corresponds to what Voigt calls 'Dehnungs coefficient' and the reciprocal of the rigidity modulus also expressed in grams weight per square millimeter is termed the deformability corresponding to what Voigt calls 'Drillungs coefficient'.

TABLE III

Young's Modulus of M Margaritifera in the plane of the shell

No	Inclination to lines of growth	Young's modulus in dynes/cm ²	Young's modulus in gm/mm ²	Extensibility
1	0°	9.25 × 10 ¹¹	9.46 × 10 ⁸	1.06 × 10 ⁻⁷
2	15°	7.82	8.00	1.25
3	30°	7.38	7.55	1.33
4	45°	8.06	8.24	1.21
5	60°	7.01	7.16	1.40
6	75°	6.06	6.20	1.61
7	90°	5.74	5.87	1.70
8	105°	6.64	6.79	1.47
9	120°	7.23	7.39	1.35
10	135°	7.37	7.54	1.32
11	150°	7.59	7.76	1.29
12	165°	9.66	9.88	1.01

TABLE IV

Young's Modulus of M Margaritifera in the transverse section

No	Inclination to the c-axis	Young's modulus in dynes/cm ²	Young's modulus in gm/mm ²	Extensibility
1	0°	2.12 × 10 ¹¹	2.17 × 10 ⁸	4.61 × 10 ⁻⁷
2	30°	1.29	1.32	7.59
3	45°	1.25	1.28	7.80
4	60°	2.81	2.87	2.46
5	75°	3.23	3.30	3.03

TABLE V

Rigidity modulus of M. Margaritifera in the plane of the shell

No	Inclination to lines of growth	Rigidity modulus in dynes/cm ²	Rigidity in gm/mm ²	Deformability
1	0°	2.66 × 10 ¹¹	2.72 × 10 ⁷	3.68 × 10 ⁻⁷
2	15°	2.62	2.68	3.73
3	30°	2.52	2.58	3.88
4	45°	2.25	2.30	4.35
5	60°	2.39	2.11	4.09
6	75°	2.10	2.15	4.66
7	90°	2.11	2.19	4.57
8	105°	2.16	2.21	4.53
9	120°	2.42	2.47	4.04
10	135°	2.36	2.11	4.15
11	150°	2.40	2.15	4.08
12	165°	2.73	2.81	3.58

TABLE VI

Elastic modulus of Turbo in the plane of the shell

No	Inclination to lines of growth	Young's modulus in dynes/cm ²	Extensibility	Rigidity modulus in dynes/cm ²	Deformability
1	0°	6.84 × 10 ¹¹	1.43 × 10 ⁻⁷	2.35 × 10 ¹¹	4.16 × 10 ⁻⁷
2	30°	6.84	1.43	2.11	4.06
3	60°	6.89	1.42	2.36	4.15
4	90°	6.43	1.52	2.48	3.94
5	120°	6.68	1.46	2.40	4.08
6	150°	6.79	1.44	2.38	4.11

TABLE VII

Young's modulus of Nautilus Pompilius in the plane of the shell

No	Inclination to lines of growth	Young's modulus in dynes/cm ²		
		1st Shell	2nd Shell	Average
1	0°	4.32×10^{11}	4.44×10^{11}	4.38×10^{11}
2	7.5°	2.31	3.41	3.34
3	15.0°	2.78	2.90	2.84
4	22.5°	2.78	2.90	2.84
5	30.0°	3.81	3.86	3.84
6	37.5°	3.72	3.86	3.79
7	45.0°	2.98	3.09	3.04
8	52.5°	4.47	4.61	4.54
9	60.0°	2.86	2.77	2.82
10	67.5°	3.02	3.13	3.08
11	75.0°	3.10	3.00	3.05
12	82.5°	2.41		2.41
13	90.0°	1.88	1.98	1.93

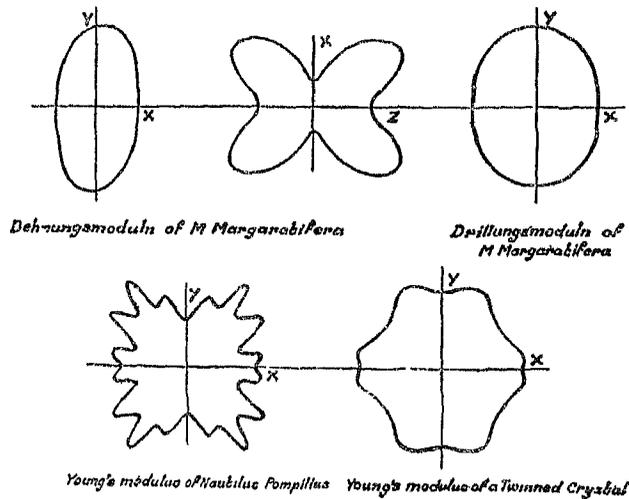


FIG III

YOUNG'S MODULUS OF CALCIFIC SHELLS

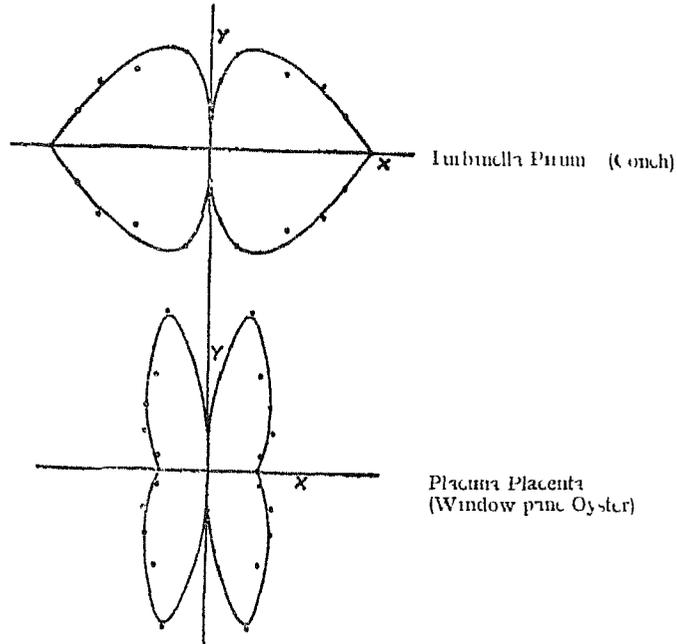


FIG. IV

TABLE VIII

Elastic Modulus of chank or 'Turbinella Pirum' in the plane of the shell

No	Inclination to lines of growth	Young's Modulus in dynes/cm ²	Extensibility
1	0°	5 58 × 10 ¹¹	1 75 × 10 ⁻⁷
2	15°	4 81	2 03
3	30°	4 50	2 17
4	45°	3 74	2 62
5	60°
6	75°	1 05	2 89
7	90°	1 05	9 32

TABLE IX

Elastic Modulus of window-pane oyster in the plane of the

No	Inclination to lines of growth	Young's Modulus in dynes/cm ²	Extensibil
1	0°	1.80 × 10 ¹¹	5.43 × 1
2	15°	1.84	5.32
3	30°	2.57	3.81
4	45°	3.13	3.13
5	60°	3.70	2.64
6	75°	5.65	1.73
7	90°	1.34	7.30

TABLE X

Young's modulus of other shells in the plane of the she

No	Shell	Young's modulus along lines of growth	Young's mo across line growth
1	M. Vulgaris	4.26 × 10 ¹¹ dynes/cm ²	.
2	Trochus	3.37 × 10 ¹¹ „	3.31 × 10 ¹¹ dyn
3	Heliotis (Abalone)	4.36 × 10 ¹¹ „	3.27 × 10 ¹¹

NOTE —The Young's modulus normal to the lines of growth has not been determined in the case of M. Vulgaris since the shell was so small that a large curvature

5 RELATION BETWEEN STRUCTURE AND ELASTICITY

A MOTHER-OF-PEARL

An analysis of the results given in the previous section brings out prominently some interesting facts and an attempt is made to correlate those facts with the known structural details, reserving a detailed consideration of individual shells to a later stage

It will be noticed that the elastic properties show a more or less regular variation with direction in the case of shells of the Lamellibranchs wherein we know from independent evidence there is a definite orientation of the crystals, while in the Gastropods where there is random orientation in the plane of the shell, the directional variation of elasticity is irregular and within the limits of experimental errors and structural faults (In HELIOTIS C, however, there is evidence of a definite though small elastic anisotropy, a fact agreeing with Ramaswami's observation that in this shell there is an arrangement similar to that in M VULGARIS but with a large error of orientation varying from 60 to 90°) In NAUTILUS where an intermediate behaviour will be expected, a totally peculiar variation is observed and it will be considered later on. Again within the same zoological group it is observed that the elasticity varies considerably from shell to shell. Thus the Young's modulus of M MARGARATIFERA along the lines of growth is 9.3 while the corresponding value for M VULGARIS is 4.3. Among the Gastropods, TURBO possesses a value of 6.5 for its Young's modulus which it is about 3.3 and 4.3 for TROCHUS and HELIOTIS C, respectively. The absolute magnitude of the elastic modulus of any shell is also of interest. Thus in M MARGARATIFERA where the orientation is of a high order of regularity, the Young's modulus in any direction is considerably less than that of aragonite in a corresponding direction. The average value of Young's modulus for a random distribution of aragonite will be about 9.3 while the actual value for TURBO in which a completely random orientation is known to exist is in the whereabouts of 6.5.

That the elastic anisotropy of the shells depend upon the

degree of orientation of the aragonite crystals in the plane of shell is a necessary consequence of the fact that the aragonite crystal itself possesses marked elastic anisotropy. Hence the orderly disposition of the crystals will more or less tend to preserve the anisotropy, while a quite random orientation will average the differences to a mean value. Comparing Figs I & III we see that the elastic curves of aragonite in the xy-plane and of shell 'M Margaritifera' in the plane of the shell are quite similar (except for the ratio of the axes) and that the maximum value of Young's modulus of aragonite occurs along its a-axis and of the shell nearly along the lines of growth. From this we infer that in 'M Margaritifera' the aragonite crystals are oriented in the plane of the shell with their a-axis, more or less, along the lines of growth.

Still the fact that the actual elasticity is considerably less than the probable value for the distribution concerned calls for an explanation. We know that in all these shells, though the bulk of the matter consists of aragonite crystals, the crystal particles themselves are bound together in a framework of conchyolin. It is also known that conchyolin possesses a very low elasticity of the order of 0.3×10^{11} dyn/cm² and that the resulting elasticity of a mixture of two substances will have an intermediate value. The drop in the elasticity of the shells is therefore to be attributed to the effect of the binding material. This assumption gets further support from the fact that within the family, the drop in the elasticity increases with the conchyolin content of the shells, as is seen from the following table.

TABLE XI

Relation between Young's modulus and Conchyolin content

No	Family	Shell	Percentage by weight of conchyolin	Young's modulus along lines of growth
1	Lamelli-branches	M Margaritifera	4.2 — 5.0	9.3×10^{11} dynes/cm ²
2	„	M Vulgaris	10.0 — 11.0	4.3 „
3	Gastropods	Trochus*	2.5 — 3.0	3.4 „
4	„	Turbo	6.5 — 7.5	6.5 „
5	„	Heliotis (Abalone C)	8.0 — 9.0	4.3 „
6	Cephalopods	Nautilus Pompilius	14.0 — 15.0	4.5 „

* Trochus appears to be an exception but may not be really so. The low value of the Young's modulus in spite of low conchyolin content might be due to want of parallelism between the surface of the test-piece and the plane of laminations, a difficulty that has already been referred to. As seen from table IV, even a small inclination to the lamination plane brings the Young's modulus down from a value 9.3 in the plane of the shell to 3.3 for an inclination of only 15° therefrom.

B CALCITIC SHELLS

The elastic modulus of the calcitic shells, chank and Placuna Placenta shows a marked anisotropy in the plane of the shell itself. This is interesting from two points of view. Ordinarily in aragonitic shells, the Gastropod family shows a random distribution of the crystals in the plane of the shell resulting in a uniform value of Young's modulus in different directions. The chank which belongs to the Gastropod family, however, shows a definite anisotropy in the plane of the shell which clearly rules out a random orientation. The next interesting point is that, the elastic properties of calcite in a plane perpendicular to the optic axis being isotropic, the large elastic anisotropy of the calcitic

shells in the plane of the shell definitely indicates an inclination of the c -axis to the plane of the shell, differing appreciably from 90° . Independent evidence of this conclusion is available both from a study of the microscopic structure and magnetic anisotropy of the shells.

Also a comparison of the values of extensibility of the shells with the corresponding values of calcite given by Voigt shows that the plane of the shell in the case of the chank corresponds to the xz plane of calcite with the x -axis parallel to the lines of growth and the z -axis in a plane perpendicular to them. Further, the large difference of the extensibility at right angles to the lines of growth in the plane of the shell suggests that the inclination of the z -axis to the plane of the shell is not very large, probably of the order of about 30° .

A similar comparison in the case of *Placuna Placenta* indicates that the plane of the shell corresponds to the yz plane of calcite with the lines of growth parallel to the z -axis. The plane of the shell however cannot be the yz plane since the microscopic as well as the magnetic evidence definitely establishes that the z -axis is inclined at an angle of 64° to the shell surface. From the close coincidence of the extensibility in the plane of the shell with that of the yz plane of calcite, we infer that the y -axis is normal to the plane of growth. Hence we infer the following facts about the disposition of the crystals in the plane of the shell: Y -axis perpendicular to the plane of growth, x -axis parallel to the lines of growth and inclined at an angle of 26° to the plane of the shell, z -axis parallel to the lines of growth and inclined at 64° to the plane of the shell*.

As in the case of the aragonitic shells, the effect of the chrysolin is to diminish the actual value of the elasticity of the shell structure in the various directions and a detailed calculation based upon the observed elastic modulus in the respective directions enables us to deduce the distribution of conchyolin in the structure.

6 'M MARGARATIFERA'

By making use of the observed values of the Young's modulus along and across the lines of growth in the plane of the shell

* Numerical value taken from Nilakantan's paper

combination with the microscopic data on the size of the crystalline units, it is possible to calculate the distribution of conchyolin in the structure. Considering the nature of the quantities involved in the calculation and the unavoidable approximations introduced therein, the results are at best only near the truth but their great claim to consideration lies in the fact that the value of the Young's modulus perpendicular to the plane of the shell deduced from the results of the calculation agrees remarkably well with the experimental value. Indeed, it was the startlingly low value of the Young's modulus in this direction according to the calculations that was the incentive for the rather difficult experimental determination of this quantity and it is gratifying to note that the trouble proved to be worth taking.

A detailed calculation is given for the shell 'M Margaritifera' since only in this case all the necessary optical and other data were available. The architecture of this shell can be compared to a brick-work on a microscopic scale wherein the particles of aragonite form the bricks and the conchyolin forms the mortar. Assuming the platelets of aragonite to be approximately rectangular, let its dimensions be a , β and γ along the crystallographic axes a , b and c and let us choose the axes of co-ordinates, x , y and z respectively parallel to a , b and c . If we assume that the conchyolin surrounding the platelet all round has got a thickness δa along a , $\delta \beta$ along b and $\delta \gamma$ along c , the unit of structure can be taken as a platelet of aragonite with adjoining conchyolin layers on one set of three mutually perpendicular faces. In effect the structural unit becomes a brick of dimensions $(a + \delta a)$, $(\beta + \delta \beta)$ and $(\gamma + \delta \gamma)$, the $a\beta\gamma$ portion of which consists of aragonite and the $\delta a - \delta \beta - \delta \gamma$ portion consists of conchyolin. Since the entire shell consists of a packing of these structural units, the Young's modulus of the shell in any direction will be the same as that of the structural unit in that direction. Hence we shall now proceed to deduce a general expression for the Young's Modulus of the structural unit in any direction.

Let q_a , q_b and q_c be the Young's modulus of aragonite along the a , b and c axes respectively and let q be the Young's modulus of

conchyolin assumed isotropic and let q_x , q_y and q_z be required values of the Young's modulus of the structural unit along the x, y and z-axes respectively

In order to find the total effect in any direction of all the conchyolin layers, we shall consider the effect first of the layer normal to that direction and then of the layers parallel to that direction. In order to find the Young's modulus q_x we shall first consider the effect of the layer of thickness δa on the aragonite and then the effect of the layers $\delta\beta$ and $\delta\gamma$. If a stress of magnitude S acts on the structural unit parallel to the x-axis,

Strain along the x-axis of the aragonite portion $= \frac{S}{q_a}$ and strain along the x-axis of conchyolin portion $= \frac{S}{q}$

The corresponding extensions are $\frac{S}{q_a} a$ and $\frac{S}{q} \delta a$ respectively

$$\text{Total strain } \epsilon = \frac{\frac{S}{q_a} a + \frac{S}{q} \delta a}{a + \delta a}$$

$$\text{Modulus } q' = \frac{S}{\epsilon} = \frac{a + \delta a}{\frac{a}{q_a} + \frac{\delta a}{q}} = \frac{q_a q (a + \delta a)}{q a + q_a \delta a}$$

Next to find the effect of $\delta\beta$ and $\delta\gamma$ on q' consider a section of the structural unit normal to the x-axis. The thickness of the conchyolin layer in the ab-plane is $\delta\gamma$ and that in the ac-plane is $\delta\beta$. Let us consider a uniform linear strain, e along the x-axis

Force on the aragonite face along the x-axis $= eq' \beta \gamma$

Force along the x-axis on the conchyolin layer of thickness, $\delta\gamma$ $\left. \vphantom{\text{Force}} \right\} = eq(\beta + \delta\beta)\delta\gamma$

Force along the x-axis on the conchyolin layer of thickness, $\delta\beta$ $\left. \vphantom{\text{Force}} \right\} = eq\gamma\delta\beta$

Total force along the x-axis $= (q' \beta \gamma + q \delta \gamma \delta \beta + q \gamma \delta \beta) e$

(In this expression and in the sequel, products of δa , $\delta\beta$, $\delta\gamma$ among themselves are neglected as quantities of the second order of magnitudes)

Final modulus along the x-axis = q_x

$$= \frac{(q'\beta\gamma + q\beta\delta\gamma + q\gamma\delta\beta)e}{(\beta\gamma + \beta\delta\gamma + \gamma\delta\beta)e} \quad (2)$$

Substituting for q' in 2 from equation 1,

$$q_x = \frac{\frac{\beta\gamma q_a q(a + \delta a)}{q_a + q_a \delta a} + q\beta\delta\gamma + q\gamma\delta\beta}{\beta\gamma + \beta\delta\gamma + \gamma\delta\beta}$$

$$= \frac{q(a\beta\gamma q_a + \beta\gamma\delta a q_a + a\beta\delta\gamma q + \gamma a\delta\beta q)}{q(a\beta\gamma + a\beta\delta\gamma + \gamma a \delta\beta) + \beta\gamma\delta a q_a} \quad (3)$$

By a cyclic variation of a , β and γ , we obtain,

$$q_y = \frac{q(a\beta\gamma q_b + \gamma a\delta\beta q_b + \beta\gamma\delta a q + a\beta\delta\gamma q)}{q(a\beta\gamma + \beta\gamma\delta a + a\beta\delta\gamma) + \gamma a\delta\beta q_b} \quad (4)$$

$$q_z = \frac{q(a\beta\gamma q_c + a\beta\delta\gamma q_c + \gamma a\delta\beta q + \beta\gamma\delta a q)}{q(a\beta\gamma + \gamma a\delta\beta + \beta\gamma\delta a) + a\beta\delta\gamma q_c} \quad (5)$$

Now of the three quantities q_x , q_y and q_z given by equations 3, 4 and 5, q_x and q_y have been determined experimentally and hence to solve for δa , $\delta\beta$ and $\delta\gamma$ another equation connecting them with known quantities is necessary and this is obtained from consideration of the percentage composition of the shells. Considering the structural unit of dimensions $(a + \delta a)$, $(\beta + \delta\beta)$ and $(\gamma + \delta\gamma)$

Volume of aragonite = $a\beta\gamma$ and

Volume of conchyolin = $(\beta\gamma\delta a + \gamma a\delta\beta + a\beta\delta\gamma)$

We know from the chemical analysis of 'M. Margaritifera' that it contains 95 per cent by weight of aragonite and 5 per cent by weight of conchyolin

Hence in 100 gm of the shell,

Weight of aragonite = 95 gm and

Weight of conchyolin = 5 gm

Density of aragonite = 2.92 gm / c.c. and

Density of conchyolin = 1.25 gm / c.c.

Hence in 100 gm of the shell,

Volume of aragonite = 32.5 c.c. and

Volume of conchyolin = 4.0 c.c.

Ratio by volume of aragonite to conchyolin = $\frac{32.5}{4.0}$ But from the dimensions of the structural unit, the ratio of the volumes

$$= \frac{\alpha\beta\gamma}{(\beta\gamma\delta\alpha + \gamma\alpha\delta\beta + \alpha\beta\delta\gamma)} = \frac{32.5}{4.0} \quad (6)$$

From a careful study of very thin sections of the shell, under the microscope, the platelets are found to have dimensions very nearly in the ratio of 3 : 10 : 1 along the a, b and c-axes respectively. Hence, in our equations we can substitute for α , β and γ the quantities 3, 10 and 1 respectively on some arbitrary unit. To get an idea of this arbitrary unit it might be remarked that the largest dimension of these platelets varies between 3 and 4μ and hence 10 of these arbitrary units make up 3 or 4μ .

Solving equations 3, 4 and 6 for $\delta\alpha$, $\delta\beta$ and $\delta\gamma$, we obtain

$$\delta\alpha = 0.0267, \quad \delta\beta = 0.095, \quad \delta\gamma = 0.1066$$

We thus find that the thickness $\delta\gamma$ of the organic material in between successive layers of aragonite is only about one-tenth the thickness of the aragonite layer itself, a fact which confirms the views of Schmidt and Raman. Further, compared to the thickness of aragonite in corresponding directions, there is maximum protein ratio in the z-axis direction.

By substituting for $\delta\alpha$, $\delta\beta$ and $\delta\gamma$ in equation 5, the Young's modulus of the shell perpendicular to its surface comes out to be 2.28×10^{11} dynes/cm². The experimental value (refer table IV) is 2.1×10^{11} dynes/cm² and the agreement is seen to be quite good.

7. 'NAUTILUS POMPILIUS'

As mentioned previously the elastic behaviour of this shell is peculiarly interesting. This shell belongs to the Cephalopod family where the crystals of aragonite are having an intermediate degree of orientation. Hence one would have expected the elastic curve to have a shape neither so elliptic as 'M. Margaritifera' nor so circular

as 'Turbo' The actual curve, however, has got a very irregular shape with sudden variations between maxima and minima. If, in the calculation given in the previous section for the Young's modulus of 'M Margaritifera', we substitute 14 per cent of conchyolin as it exists in 'Nautilus' instead of the 5 per cent as in 'M Margaritifera', we obtain the value of Young's modulus somewhere about 4.8 and it is interesting to note that in the experiment on 'Nautilus', the value of the modulus in any direction does not exceed this limit.

From X-ray studies, Ramaswamy has pointed out that in 'Nautilus' the crystals of aragonite are twinned across the plane 110. A group consisting of two pairs of oppositely directed twins is necessary to explain the X-ray pattern. From the point of view of elasticity, however, the two pairs are identical since the elastic curve of aragonite is symmetrical with respect to the axes. Assuming for a moment that a similar twinning were to take place in 'M Margaritifera' we can easily calculate the effect on the elastic curve of this twinning. For finding the Young's modulus in any direction of a twinned structure, we shall have to compound in that direction the effect of the two crystals inclined to each other at an angle of 120° . If q_1 and q_2 represent the Young's modulus along any direction due to the two components of a twinned structure, the resultant value q in that direction will be

$$\frac{2 q_1 q_2}{q_1 + q_2}$$

The following table gives the calculated values of the Young's modulus for a twinned structure

TABLE XII

Young's modulus of a twinned structure

No	Inclination to lines of growth	q_1	q_2	q
1	0°	7.38	7.38	7.38
2	15°	8.06	7.82	7.94
3	30°	7.01	9.25	7.92
4	45°	6.06	7.82	6.83
5	60°	5.74	7.38	6.46
6	75°	6.64	8.06	7.28
7	90°	7.23	7.01	7.12

We see from the curve (Fig III) that the effect of twinning is to considerably modify the elastic behaviour. While the elastic curve of untwinned crystals in 'M. Margaritifera' is a uniform ellipse, that of a twinned structure shows maxima and minima within the same quadrant. It should, however, be admitted that the maxima and minima of the twinned structure are fewer and much less pronounced than those representing the actual behaviour of 'Nautilus'. Twinning may be responsible to some extent for the peculiar behaviour of 'Nautilus' but it looks as though it is not the whole cause. The large quantity of conchyolin present in this shell, introduces, perhaps, complications in the elastic properties. One remarkable feature that was noticed in the present investigation was that, of all the shells examined, it was only 'Nautilus' that showed an elastic hysteresis to a well-defined and large extent. Possibly the distribution of conchyolin in the plane of the shell is of such a character as to give rise to the sudden variation in elastic properties from direction to direction. When thin sections of this shell are examined between crossed nicols

under the polarising microscope, the conchyolin appears as dark patches and the size and distribution of these dark patches are found to be of a complex character, a fact which lends further support to this idea

8 PLACUNA PLACENTA

By the method employed in the case of *M Margaritifera* for elucidating the conchyolin distribution in the shell it is possible to do the same for this shell. The values of the extensibility of calcite in the plane of the shell parallel and perpendicular to the lines of growth and in a direction perpendicular to the plane of the shell have been obtained by substituting proper values of the direction cosines l , m and n in the general expression given by Voigt for the extensibility of calcite in any direction. Remembering that the extensibility is the reciprocal of the Young's modulus expressed in grams weight per sq mm, the corresponding values of Young's modulus in dynes per sq cm have been calculated. The general expression for the extensibility $E(l, m, n)$ in a direction whose direction cosines with x , y and z -axes are respectively l , m and n is $E(l, m, n) =$

$$11.14(1-n^2)^2 + 17.13n^4 + 31.05m^2n^2 + 17.97mn(3l^2 - m^2)$$

The direction of the lines of growth in the plane of the shell corresponds to a direction in the XZ plane of calcite inclined at an angle of 64° to the z -axis and 26° to the x -axis, while the direction at right angles to the lines of growth corresponds to the y -axis of calcite. The direction normal to the plane of the shell corresponds to a direction in the xz -plane of calcite inclined at an angle of 26° to the z -axis and 64° to the x -axis. Hence the extensibility corresponding to the three directions will be obtained by substituting

$$\begin{aligned} & l = \cos 26, m = 0 \text{ and } n = \cos 64, \text{ for the first direction,} \\ & l = n = 0 \text{ and } m = 1, \text{ for the second direction and} \\ & l = \cos 64, m = 0 \text{ and } n = \cos 26 \text{ for the third direction} \end{aligned}$$

Thus calling the three directions a , b and c , we obtain

$$\begin{aligned} E_a &= 7.901 \times 10^{-8} \text{ and } q_a = 12.38 \times 10^{11} \text{ dynes per sq cm} \\ E_b &= 11.14 \times 10^{-8} \text{ and } q_b = 8.78 \times 10^{11} \text{ ,, ,,} \\ E_c &= 11.61 \times 10^{-8} \text{ and } q_c = 8.74 \times 10^{11} \text{ ,, ,,} \end{aligned}$$

From the experiment, we know that $q_x = 1.8 \times 10^{11}$ dynes/cm²
and $q_y = 1.34 \times 10^{11}$ dynes/cm²

The percentage of calcite in the shell = 89.7 and of conchyolin is 10.3

Density of calcite = 2.72 gms / c.c. and

Density of conchyolin = 1.25 gms / c.c.

In 100 gms of shell,

$$\text{volume of calcite} = \frac{89.7}{2.72} = 32.98 \text{ c.c.}$$

$$\text{and } \text{,, } \text{,, conchyolin} = \frac{10.3}{1.25} = 8.25 \text{ c.c.}$$

$$\left. \begin{array}{l} \text{Ratio of the volume of calcite} \\ \text{to conchyolin} \end{array} \right\} = \frac{32.98}{8.25} = 4.0$$

The values given by Schmidt for the platelets of calcite in Placuna Placenta are $100\mu \times 5\mu \times 1\mu$

$$a = 100\mu, \beta = 5\mu \text{ and } \gamma = 1\mu$$

Substituting these values in equations 3, 4 and 6 and solving for δa , $\delta \beta$ and $\delta \gamma$ we obtain $\delta a = 16.9\mu$, $\delta \beta = 1.22\mu$ and $\delta \gamma = 0.163\mu$

The values of the conchyolin thickness surrounding the calcite crystallite obtained above indicate a very liberal distribution of the protein, particularly the thickness of conchyolin along the lines of growth between neighbouring platelets of calcite comes out to be of the order of nearly 17μ about a sixth of the length in this direction of the calcite platelet. Without being actually present in such large patches, an equivalent effect could be produced by even a small error of orientation of the crystallites. Since the crystallites are very long compared to their cross-dimensions, even an error of orientation of 5° will make the effective thickness of conchyolin along the lines of growth very large. Since it is known that there is a certain degree of error of orientation in this shell, the high value of δa is very likely the effect of this fact.

A comparison of the relative thicknesses of crystal to protein in 'M. Margaritifera' and Placuna Placenta is very significant

	∂a a	$\delta\beta$ β	$\delta\gamma$ γ
M Margaritifera	0 0089	0 0095	0 107
Placuna Placenta	0 16	0 24	0 16

It is seen at once that the conchyolin distribution is more or less uniform all round in Placuna Placenta while in M Margaritifera it occurs much more liberally normal to the plane of the shell than in the plane of the shell. Also in conformity with the greater conchyolin content of this shell, Placuna Placenta has a very liberal distribution of conchyolin all round. This explains both the easy flaking of this shell and its easy tearing in the plane of the flakes. Evidently also the low values of the elastic modulus are due to this large protein content.

9 SUMMARY AND CONCLUSION

The elastic properties of mother-of-pearl obtained from a number of molluscan shells have been determined in different directions with respect to the lines of growth. In the case of 'M Margaritifera' the Young's modulus in the transverse section also has been determined. As a general rule it is found that in all the shells examined, the Young's modulus in any given direction diminishes with increasing protein content.

In 'M Margaritifera' there is a large elastic anisotropy in the plane of the shell, the values of the Young's modulus parallel and normal to the lines of growth being 9.3×10^{11} dynes/cm² and 5.8×10^{11} dynes/cm², the shape of the elastic curve being similar to that of aragonite in the XY-plane. The torsional properties in the plane of the shell show also an anisotropy similar to aragonite in the XY-plane. These observations agree well with the known existence of orientation in the plane of the shell of the crystal particles composing the shell. The Young's modulus in the transverse section is found to have a remarkably low value.

The elastic behaviour of 'Turbo' and 'Trochus' in the plane of the shells is, on the other hand, isotropic agreeing well with the known randomness of crystal orientation present in these shells. In

'*Heliotis* (abalone) there is evidence of a definite though small elastic anisotropy, as is to be anticipated from the X-ray observations on this shell

A general expression for calculating the elastic modulus of a compound structure in terms of the elastic moduli of component materials and their distribution is derived and by the use of this expression in combination with the observations on the Young's modulus of '*M. Margaritifera*', the distribution of conchyolin in this shell is deduced. The formulæ indicate, in agreement with other evidence that the conchyolin layers between the aragonite layers are very thin in comparison with the latter. The low value of the Young's modulus in the direction normal to the laminations as found by the experiment is also explained by the theory.

The elastic behaviour of '*Nautilus Pompilius*' is found to be very peculiar, showing rapid variations in the value of the Young's modulus from direction to direction in the plane of the shell. Though twinning which is known to be present in this shell, might account for this to some extent, the real cause is probably to be traced to some peculiarity in the distribution of the large amount of conchyolin present in this shell.

The elastic behaviour of the calcitic shells is found to be in conformity with the known inclination of the c-axis of calcite to the plane of the shells and an estimate of the protein distribution in *Placuna Placenta* has been deduced from considerations of its elastic behaviour. The protein thickness all round comes out to be quite large particularly in the direction of the lines of growth and a possible alternative view of this fact based upon the known error of orientation of the crystallites is suggested.

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